

PCTWORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : B01J 2/30, A23K 1/02, A23P 1/06	A1	(11) International Publication Number: WO 98/53907 (43) International Publication Date: 3 December 1998 (03.12.98)
(21) International Application Number: PCT/FI98/00449 (22) International Filing Date: 28 May 1998 (28.05.98) (30) Priority Data: 972264 28 May 1997 (28.05.97) FI (71) Applicant (for all designated States except US): CULTOR CORPORATION [FI/FI]; Kyllikinportti 2, FIN-00240 Helsinki (FI). (72) Inventors; and (75) Inventors/Applicants (for US only): MOILANEN, Raija [FI/FI]; Taimentie 6 as. 4, FIN-20760 Piispanristi (FI). HALLANORO, Helena [FI/FI]; Apilakatu 1, FIN-21110 Naantali (FI). (74) Agent: KOLSTER OY AB; Iso Roobertinkatu 23, P.O. Box 148, FIN-00121 Helsinki (FI).		(81) Designated States: AL, AM, AT, AT (Utility model), AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, CZ (Utility model), DE, DE (Utility model), DK, DK (Utility model), EE, EE (Utility model), ES, FI, FI (Utility model), GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (Utility model), SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>In English translation (filed in Finnish).</i>
(54) Title: BETAINE PRODUCT, METHOD FOR ITS MANUFACTURE, AND ITS USE (57) Abstract The invention relates to a solid betaine product having good treatment properties. The invention also relates to a method for the manufacture of the betaine product and its use, as well as a feed containing said betaine product and other feed components. Said solid betaine product contains a fluidity improver and/or anti-agglomeration agent, and a hydrophobic, moisture-proof layer is arranged on the surface of the particles.		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

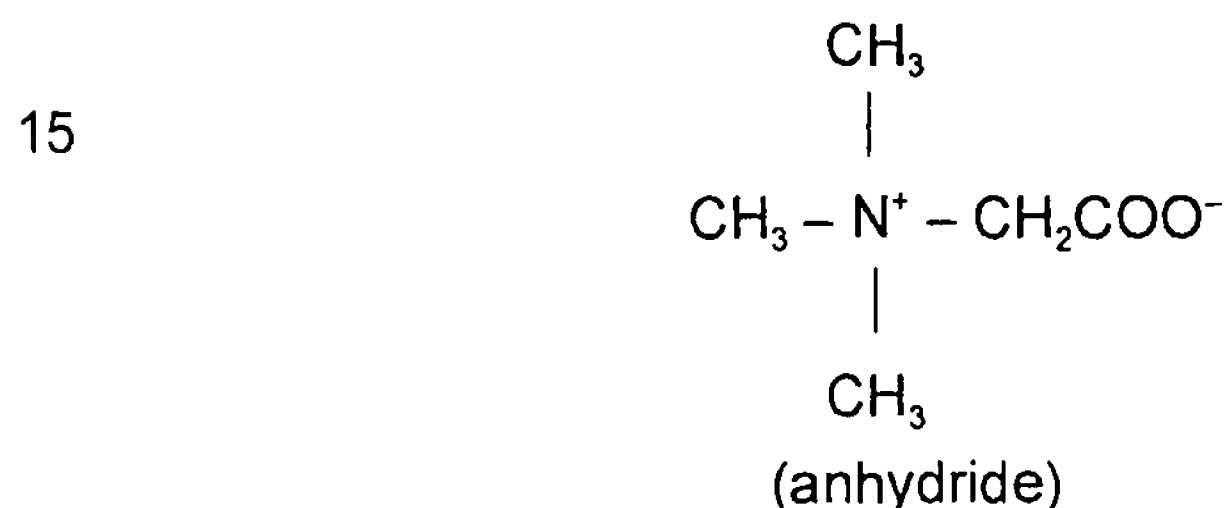
BETAINE PRODUCT, METHOD FOR ITS MANUFACTURE, AND ITS USE

FIELD OF THE INVENTION

The invention relates to a solid betaine product having good treatment properties. The invention also relates to a method for the manufacture of the betaine product, and its use, as well as an animal feed containing said betaine product and other feed components.

BACKGROUND OF THE INVENTION

Betaines are fully N-methylated amino acids. Betaines are natural products that have an important function in both plant and animal metabolism. One of the most common betaines is a glycine derivative in which three methyl groups are bonded to the nitrogen atom of the glycine molecule. This betaine compound is usually called betaine, glycinebetaine or trimethylglycine, and it has the following structural formula:



Other betaines include, for example, alaninebetaine and prolinebetaine, which has been reported, among other things, to prevent osteomalacia in chickens. A detailed description of betaines is given by R.G. Wyn Jones and R. Storey in *The Physiology and Drought Resistance in Plants*, ed. L.G. Paleg and D. Aspinall, Academic Press, Sydney, Australia, 1981.

Betaine thus has a bipolar structure and it contains several chemically reactive methyl groups, which it can donate in enzyme-catalysed reactions. Most organisms are able to synthesize small quantities of betaine e.g. for the methyl function, but they are not able to react to stress by substantially increasing the production and storage of betaine.

Betaine is a highly hygroscopic substance, and therefore its treatment and use as such or as part of an end product causes problems in humid conditions, since it turns viscous, lumpy and poorly flowable. In order to treat and use betaine without problems, it is necessary to protect it against humidity. Hydrophilic fluidity improvers / anti-agglomeration agents (which absorb

free water) have been used previously. However, hydrophilic substances of this kind are not able to protect betaine in humid conditions, even though they improve the fluidity of betaine in good, dry conditions.

Betaine is used, among other things, as a feed additive and as a
5 crop improver of plants under stress conditions, as well as in cosmetic, pharmaceutical and food industries.

Betaine can be obtained, for example, from sugar beet by chromatographic methods. Betaine is commercially available e.g. under the registered trademark BETAFIN[®], Cultor Oy, Finnsugar Bioproducts. BETAFIN[®] is solid
10 betaine (betaineanhydride or betainemonohydrate) produced by Finnsugar Bioproducts.

EP 0 573 876 discloses a method for the manufacture of a product containing cholinechloride by mixing cholinechloride solution in fine drops with a pulverized, hydrophobic substance, and by drying the mixture with a fluidized bed drier. Silica made hydrophobic by silanization or a metallic salt of a
15 fatty acid can be used as the hydrophobic substance.

BRIEF DESCRIPTION OF THE INVENTION

Now it has been found that by means of hydrophobic substances a moisture-proof layer can be arranged on the surface of betaine particles.
20 Hence fluidity and anti-agglomeration of a betaine product, its use as such or as a part of the end-product, can substantially be improved also in humid conditions. Hydrophobic substance is preferably added to a betaine product by utilizing an efficient blender.

DETAILED DESCRIPTION OF THE INVENTION

25 The invention relates to a solid betaine product which contains a fluidity improver and/or anti-agglomeration agent. In accordance with the invention, a hydrophobic, moisture-proof layer is arranged on the surface of particles.

The solid betaine product refers to a crystalline or ground product.
30 The particle size of the crystalline product is 0.05 to 2.0 mm, preferably on the average about 0.3 to 0.6 mm. Preferably, the crystalline product does not contain practically any dustlike substance, i.e. particles with the diameter of less than about 0.01 mm. The particle size of the ground product is 1 to 100 μm .

The hydrophobic layer on the surface of the particles may consist of a metallic salt of a fatty acid, such as calcium or magnesium stearate, or hydrophobic silica. Additionally, a substance which enhances said hydrophobic substance to adhere to or spread onto the surface of betaine particles may be present on the surface of the crystals.

Suitable substances that enhance the adhesion or spreading of the hydrophobic substance comprise fats, oils, fat-like substances, such as lecithins and waxes, and water-insoluble substances, such as cellulose derivatives and silicone derivatives.

In accordance with the invention, a new betaine product can be produced, for example, by mixing the blend of betaine and a hydrophobic, moisture-proof substance.

In the method of the invention, various mixers can be used. When only powdery hydrophobic substance is used, suitable mixers comprise various batch mixers or continuous mixers, such as drum mixers or helical/screw mixers. Mixing time depends on the power of the mixer, and also on the batch size. The object is that the hydrophobic substance mixes as evenly as possible with the betaine. In the ideal case, the powder forms a monomolecular layer on the surface of the product. Preferably, the smallest betaine crystals are screened out before a treatment with the hydrophobic substance.

When a binding agent is added to the powdery hydrophobic substance, spray granulators or fluidized bed drying/coating devices can be used in the method of the invention, in addition to various batch mixers and continuous mixers.

Various grinders, such as an air jet grinder or a turbo mill or some other mill useful for grinding sugar crystals, can be used in the method of the invention. The betaine is thus ground with the powdery hydrophobic substance, and the obtained powdery product flows freely and it will not agglomerate.

In accordance with one preferred method of the invention, melted, hot (e.g. about 40 °C higher than the melting point) fat is first sprayed onto the surface of the betaine crystals. It is preferable to use a hydrogenated fat with a high melting point, whereby a product is obtained, which retains good fluidity (does not become viscous) even at tropical temperatures. Other fats/oils can also be used (depending on the use of the product). When using oils that are fluid at room temperature, no heating is needed. When using fats with a high

melting point, it is preferable to preheat the betaine crystals close to the melting point of the fat. This facilitates the forming of a layer as even as possible. Thereafter the surface of the warm crystal coated with fat is 'powdered' with a hydrophobic substance. Managing the processing conditions is critical
5 (temperatures) in order that even and comprehensive protection is achieved, and the product does not stick to the walls of the equipment or elsewhere, and the product remains single, flowable crystals (no agglomeration).

Instead of spraying the fat, another technique may also be used, in which the betaine particles and the solid fat flakes, et cetera, are first mixed
10 together. Thereafter, the blend is heated to exceed the melting point of the fat while it is continuously being mixed.

The fat treatment may be carried out by using a mixer with the possibility of adding liquid (spray nozzle systems) or a spray granulator or devices of the type of fluidized bed drier/coater.

15 Useful hydrophobic substances include, for example, the following:

Metallic salts of long-chain fatty acids:

Stearates:

metallic salts of stearic acid in powder form,
(melting point in excess of 150 °C)

20 e.g. Ca-stearate, Ceasit Leviss, manufactured by Bärlocher, Germany

Ca-stearate CPR-2, manufactured by Akcros Chemicals V.O.F.,
Holland

Silica-based hydrophobic substances:

25 e.g. Sipernat D17, manufactured by Degussa Ltd., Germany

Aerosil R972, manufactured by Degussa Ltd., Germany

(and other hydrophobic Sipernat and Aerosil products)

Cellulose derivatives:

e.g. Ethocel, manufactured by Dow USA (in powder form)

30

Useful substances that enhance the adhesion or spreading of the hydrophobic substance include, for example, the following:

Fats:

35 Revel S (acetylated monoglyceride, hydrogenated soy oil); melting point 63 °C; manufactured by Lodders Croklaan, Holland

Revel C; melting point 58 °C,

Revel F; melting point 46 °C,
Revel AC; melting point 37 °C,
Dynasan P 60; manufactured by Huls AG, Germany

Oils:

5 Fish oils

Lecithins:

e.g. lecithins of Metarin product line; manufactured by Lucas
Mayer, Germany

Silicone derivatives (spraying in solution):

10 e.g. Pharsil MK silicone emulsion; manufactured by: Wacker Chemie GmbH, Germany

Cellulose derivatives (spraying in solution):

e.g. Ethocel; manufactured by Dow USA

15 **Example 1**

Betaine anhydride (Nutristim, manufactured by Cultor Oy; quantity 50 kg, granule size 0.27 to 1.0 mm) was introduced into a drum mixer (Forberg F-60, manufactured by Fa. Halvor Forberg A/S, Norway) and 3 kg calcium stearate (Ceasit Leviss, manufactured by Bärlocher, Germany, particle size 20 99% < 71 µm) was added thereto. Mixing was carried out at room temperature for about three minutes. The obtained product had good fluidity, and it remained fluid for at least three hours at a temperature of 30 °C, the relative air humidity being 95%.

Example 2

25 Into a continuous screw mixer (length of the mixing worm 4 m, diameter 400 mm, two mixing points in the screw, manufactured by Siirtoruuvi Oy, Finland) were fed betaine anhydride (Nutristim, manufactured by Cultor Oy; granule size 0.27 to 1.0 mm) 5,000 kg/hour and 6% calcium stearate (Ceasite Leviss, manufactured by Bärlocher, Germany; particle size 30 99% < 71 µm) through an additive dispenser (Accurate, manufactured by Accurate Dry Material Feeders Inc., USA). Mixing was carried out continuously in the mixing helix/screw. Temperature was 20 °C and duration of mixing in the mixing helix was two minutes. The obtained product had good fluidity and it remained fluid at least for three hours at a temperature of 30 °C, the relative air humidity be-
35 ing 95%.

Example 3

Betaineanhydride (Nutristim, manufactured by Cultor Oy; quantity 47.5 kg, granule size 0.27 to 1.0 mm) was introduced into a drum mixer (Forberg F-60, manufactured by Fa. Halvor Forberg A/S, Norway) and pre-
5 heated to 55 °C. Thereafter 2.5 kg hot (100 °C) fat (Revel-S, manufactured by Loders Croklaan, Holland) was added by spraying (nozzle size 4004, pressure 3 bar) and simultaneously mixing (for 1.3 minutes). Finally, 1 kg calcium stearate (Ceasit Leviss, manufactured by Bärlocher, Germany; particle size 99% < 71 µm) was added, and the blend was cooled to 30 °C and mixed for
10 3.7 minutes. The obtained product's solubility in water was poor and fluidity was good: it remained fluid at least for three hours at a temperature of 30 °C, the relative air humidity being 95%.

Example 4

The test was conducted in the same way as in Example 1, but in-
15 stead of calcium stearate, hydrophobic silica (Sipernat D 17, manufactured by Degussa, Germany; average granule size 10 µm) was used. The obtained product was very fluid, and it remained fluid at least for three hours at a temperature of 30 °C, the relative air humidity being 95%.

Example 5

20 The test was conducted in the same way as in Example 3, but instead of calcium stearate, hydrophobic silica (Sipernat D 17, manufactured by Degussa, Germany; average granule size 10 µm). The obtained product's solubility in water was poor and fluidity was good: it remained fluid at least for three hours at a temperature of 30 °C, the relative air humidity being 95%.

Example 6

25 Betainemonohydrate (technical quality, manufactured by Cultor Oy; quantity 47.5 kg, granule size 0.25 to 1.25 mm) was introduced into a drum mixer (Forberg F-60, manufactured by Fa. Halvor Forberg A/S, Norway) and preheated to 55 °C. Thereafter, 1.5 kg hot (100 °C) fat (Revel-S, manufactured
30 by Loders Croklaan, Holland) was added by spraying (nozzle size 4004, pressure 3 bar) and simultaneously mixing (for 1.3 minutes). Finally, 1.5 kg calcium stearate (Ceasit Leviss, manufactured by Bärlocher, Germany; particle size 99% < 71 µm) was added, and the blend was cooled to 30 °C and mixed for 3.7 minutes. The obtained product's solubility in water was poor and fluidity
35 was good.

Example 7

The test was conducted in the same way as in Example 6, but instead of calcium stearate, hydrophobic silica (Sipernat D 17, manufactured by Degussa, Germany; average particle size 10 μm) was used. The obtained
5 product had good fluidity and it remained fluid at least for three hours at a temperature of 30 °C, the relative air humidity being 95%.

Example 8

Betaine anhydride (Betafin BP, manufactured by Cultor Oy; particle size 0.05 to 1.0 mm) was fed into a continuous air jet grinder (Pulva FP Micronization, manufactured by Oy Finnpulva Ab); simultaneously, hydrophobic
10 silica (Aerosil R 972, manufactured by Degussa; average particle size 16 nm) was also fed into the grinder. The total quantity of betaine anhydride was 400 kg and that of silica 4 kg (1%). The average grinding fineness of the obtained product was less than 20 μm , the product was very fluid and it did not agglom-
15 erate.

CLAIMS

1. A solid betaine product which contains a fluidity improver and/or anti-agglomeration agent, **characterized** in that a hydrophobic, moisture-proof layer is arranged on the surface of particles.
- 5 2. A product as claimed in claim 1, **characterized** in that the layer consists of a metallic salt of a fatty acid, such as calcium or magnesium stearate.
3. A product as claimed in claim 1, **characterized** in that the layer consists of hydrophobic silica.
- 10 4. A product as claimed in claim 1, 2 or 3, **characterized** in that a substance which enhances the hydrophobic substance to adhere or to spread is additionally arranged on the surface of the crystals.
5. A product as claimed in claim 4, **characterized** in that the substance which enhances the hydrophobic substance to adhere or to spread
15 is selected from the group of fat, oil, fat-like substances, such as lecithins and waxes, and water-insoluble substances, such as cellulose derivatives and silicon derivatives.
6. A method for the manufacture of a crystalline betaine product as claimed in claim 1, 2 or 3, **characterized** in that the solid betaine and
20 the hydrophobic substance are mixed together.
7. A method as claimed in claim 6, **characterized** in that the solid betaine is mixed with a metallic salt of a fatty acid, such as calcium or magnesium stearate.
8. A method as claimed in claim 6, **characterized** in that the
25 solid betaine is mixed with hydrophobic silica.
9. A method as claimed in claim 1, 2 or 3, **characterized** in that the betaine is ground with the hydrophobic substance.
10. A method for the manufacture of a crystalline betaine product as claimed in claim 4 or 5, **characterized** in that oil or melted fat is first ar-
30 ranged on the surface of the betaine crystals, whereafter a hydrophobic substance is arranged on the surface.
11. A method as claimed in claim 10, **characterized** in that the melted fat is a hydrogenated fat with a high melting point, and the betaine crystals are preheated close to the fat melting point.
- 35 12. A method for the manufacture of a betaine product as claimed in claim 4 or 5, **characterized** in that, first, the betaine crystals and the

solid fat are mixed together, whereafter the hydrophobic substance, such as a metallic salt of a fatty acid or hydrophobic silica, is added, and the blend is heated to exceed the fat melting point while it is continuously being mixed.

13. Use of a product as claimed in any one of claims 1 to 5 as an
5 animal feed additive.

14. Use of a product as claimed in any one of claims 1 to 5 in fertil-
izers.

15. Use of a product as claimed in any one of claims 1 to 5 in cos-
metic skin care products or in toiletries.

10 16. Use of a product as claimed in any one of claims 1 to 5 in a
pharmaceutical product or in a product of unverified pharmacological effects.

17. Use of a product as claimed in any one of claims 1 to 5 in a food
product.

15 18. An animal feed, **characterized** by containing betaine
product as claimed in any one of claims 1 to 5 and other feed components.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/FI 98/00449

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B01J 2/30, A23K 1/02, A23P 1/06
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: A23K, A23L, A23P, B01J, A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WPI/Derwent's abstract, No 75-43835W, week 7526, ABSTRACT OF JP, 438407 (AS KIRG ORG CHEM), 28 January 1975 (28.01.75), abstract	1,6,9,13,18
Y	--	2-3,7-8, 14-17
X	WPI/Derwent's abstract, No 78-83702A, week 7825, ABSTRACT OF JP, 586889 (AS KIRG ORG CHEM), 6 January 1978 (06.01.78), abstract	1,6,13,18
Y	--	2-3,7-8, 14-17

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "I" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"Z" document member of the same patent family

Date of the actual completion of the international search

1 Sept 1998

Date of mailing of the international search report

07.09.1998

Name and mailing address of the ISA
Swedish Patent Office
Box 5055, S-102 42 STOCKHOLM
Facsimile No. +46 8 666 02 86

Authorized officer
Mårten Hulthén
Telephone No. +46 8 782 25 00

INTERNATIONAL SEARCH REPORT

International application No.

PCT/FI 98/00449

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	FR 2518426 A1 (GROUPEMENT D'ETUDES ET DE RECHERCHES CHARGES), 24 June 1983 (24.06.83), claims 1,3 --	2,7
Y	US 3580519 A (KURT KLEIN), 25 May 1971 (25.05.71), claim 1, abstract --	3,8
Y	WO 9535022 A1 (COMMONWEALTH SCIENTIFIC AND INDUSTRIAL RESEARCH ORGANISATION), 28 December 1995 (28.12.95), abstract --	14
Y	WO 8102420 A1 (SUOMEN SOKERI OY), 3 Sept 1981 (03.09.81), page 1, line 11 - line 15, abstract --	15-16
Y	WPI/Derwent's abstract, No 78-26128A, week 7814, ABSTRACT OF JP 53018728 (KYOWA FERMENTATION KK), 21 February 1978 (21.02.78), abstract --	17
A	FR 2151076 A (UENO FINE CHEMICAL INDUSTRIES, LTD.), 13 April 1973 (13.04.73), example 1 -----	1-18

INTERNATIONAL SEARCH REPORT

Information on patent family members

27/07/98

International application No.

PCT/FI 98/00449

Patent document cited in search report			Publication date	Patent family member(s)			Publication date
FR	2518426	A1	24/06/83	NONE			

US	3580519	A	25/05/71	CH	500001	A	15/12/70
				DE	1642998	A	13/04/72
				FR	1555348	A	24/01/69
				GB	1213494	A	25/11/70
				NL	163969	B,C	16/06/80
				NL	6803231	A	24/09/68
				SE	339458	B	11/10/71

WO	9535022	A1	28/12/95	AU	683177	B	30/10/97
				AU	2707195	A	15/01/96
				AU	PM632294	D	00/00/00

WO	8102420	A1	03/09/81	AT	11132	E,T	15/01/85
				BE	887652	A	15/06/81
				BG	49616	A	16/12/91
				CS	256365	B	15/04/88
				CS	8101434	A	17/09/87
				DK	158222	B,C	16/04/90
				DK	453281	A	13/10/81
				EP	0054544	A,B	30/06/82
				SE	0054544	T3	
				FI	77845	B,C	31/01/89
				FI	812912	A	17/09/81
				HR	940007	A,B	29/02/96
				JP	2050895	B	05/11/90
				JP	57500286	T	18/02/82
				SI	8110505	A	30/06/97
				SU	1189334	A	30/10/85
				US	4359430	A	16/11/82

FR	2151076	A	13/04/73	AU	473667	B	01/07/76
				AU	4601372	A	07/03/74
				CA	985893	A	23/03/76
				JP	48034079	A	15/05/73
